

Thermochemical Process Heat Requirements of the Copper-Chlorine Cycle for Nuclear-Based Hydrogen Production

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Abstract

A key challenge facing the future hydrogen economy is a sustainable, lower-cost alternative to current hydrocarbon-derived technologies of hydrogen production. Nuclear-based hydrogen production with a thermochemical copper-chlorine (Cu-Cl) cycle is a promising alternative that would use nuclear heat to split water directly into oxygen and hydrogen, without indirect generation of electricity through electrolysis. Heat transfer has a paramount importance in thermal efficiency of the Cu-Cl cycle. The overall cycle includes various endothermic and exothermic reaction chambers, as well as heat exchangers that supply or recover heat to individual steps within the thermochemical cycle. Heat exchangers are typically located before and after each chemical reactor to supply fluid temperatures required for the subsequent reactions. This paper examines the heat matching between steps of the Cu-Cl cycle, in efforts to recover as much heat as possible and minimize the net heat supply to the cycle, thereby improving its overall efficiency.

1. Introduction

Manufacturing and automotive industries are vulnerable to rising oil prices, since they are dependent on petroleum products that are needed for manufacturing, component materials (such as plastics) and other purposes. Currently the world consumes about 85 million barrels of oil and 104 trillion cubic feet of natural gas per day, releasing greenhouse gases that lead to global warming. In contrast, hydrogen is a clean energy carrier. Some have questioned whether the “hydrogen economy” is for us, or far in the future for our grandkids. But the worldwide hydrogen market is already valued at over \$282 billion/year, growing at 10%/year, doubling to 20%/year by 2010, doubling again to 40%/year by 2020 and expected to reach several \$trillions by 2020. A key challenge facing this rapid growth is a sustainable route to hydrogen production. This paper examines a novel route to hydrogen production based on advanced CANDU technology. Thermochemical water decomposition driven by nuclear heat with a copper-chlorine (Cu-Cl) cycle would split water into hydrogen and oxygen through intermediate copper and chlorine compounds (Serban, Lewis, Basco; [1]). Unlike other sustainable energy resources (e.g., solar, wind), nuclear heat used in this thermochemical cycle enables large-scale direct production of hydrogen. A Cu-Cl plant could be eventually linked with Canada’s nuclear reactors to achieve higher efficiencies of hydrogen production than any other conventional technology.

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Hydrogen demand is expected to increase dramatically during the next decade. As of December 2004, the Canadian hydrogen capacity reached 2.88 million tonnes per year. Within this total, Western Canada used 2.23 million tonnes per year, Eastern Canada used 0.5 million tonnes per year and the remainder of 0.15 million tonnes per year was needed by the Atlantic Provinces. Canadian use of hydrogen is projected to reach over 6 million tonnes per year by 2023. In Alberta alone, oil sands development is requiring massive quantities of hydrogen to upgrade bitumen to synthetic crude. Anticipated hydrogen demands for upgrading of heavy oil are expected to grow from the current 0.78 million tonnes per year to 3.1 million tonnes per year, or almost 50% of total Canadian production by 2023. Unlike steam-methane reforming (SMR), which uses natural gas for hydrogen production, thermochemical methods do not emit greenhouse gases.

Hydrogen production for sustainable development was examined by Bertel [2], Duffey and Miller [3 - 5]. Kloosterman [6] has predicted that hydrogen demand from the chemical, fertilizer and petrochemical industries alone will rise by a factor of four over the next decade. This is very rapid growth, even by oil-industry standards, especially since it precedes an expected phase of far higher growth in the emerging hydrogen economy for the transportation sector. Dincer [7] has outlined the key technical and environmental concerns of hydrogen production. The recent rise in oil prices and the need to sequester CO₂ has tilted the economic balance away from the traditional SMR technology [8]. A comprehensive overview of various hydrogen production schemes was presented by Yildiz and Kazimi [9]. Rosen [10 - 11] has evaluated the thermodynamic efficiency of electrolytic hydrogen production from nuclear energy and other sources. Operating temperatures are key factors in thermochemical hydrogen production. As a result, optimization of heat flows is important for high energy conversion efficiency (Naterer [12]).

Much effort internationally is being focused on the Sulfur-Iodine (S-I) cycle and its variations, as well as High-Temperature Electrolysis (HTE) for nuclear-based hydrogen production. This cycle requires temperatures in excess of 850°C from Very High Temperature Reactors (VHTR). In Japan, a bench-top S-I cycle was completed successfully in June, 2004, involving one week of continuous hydrogen production. Current Japanese efforts are completing a pilot plant and planning a commercial nuclear-hydrogen plant, which would supply hydrogen for millions of fuel cell vehicles by the end of the next decade. JAEA (Japan Atomic Energy Agency) plans to use S-I technology to co-produce electricity and hydrogen with their HTGR reactor. The reactor's 600 MWt output would produce 175 MWe electricity, as well as 640,000 m³/day of hydrogen from the S-I cycle and fresh water through a desalination plant coupled with the reactor. The copper-chlorine (Cu-Cl) cycle offers a number of advantages over the S-I cycle, including lower operating temperatures, capability of utilizing low-grade waste heat from nuclear reactors and lower-cost materials. This article investigates the heat requirements of the Cu-Cl cycle and presents recent UOIT-led developments to build an integrated lab-scale Cu-Cl cycle, in conjunction with Atomic Energy of Canada Ltd., Argonne National Laboratory and other partners.

2. Heat Requirements of the Copper-Chlorine Cycle

The Cu-Cl thermochemical cycle uses a series of reactions to achieve the overall splitting of water into hydrogen and oxygen as follows:



The Cu-Cl cycle splits water into hydrogen and oxygen through intermediate copper and chlorine compounds. These chemical reactions form a closed internal loop that re-cycles all chemicals on a continuous basis, without emitting any greenhouse gases externally to the atmosphere. Steps in the Cu-Cl cycle with and a possible realization of the cycle are shown in Table 1 and Fig. 1, respectively.

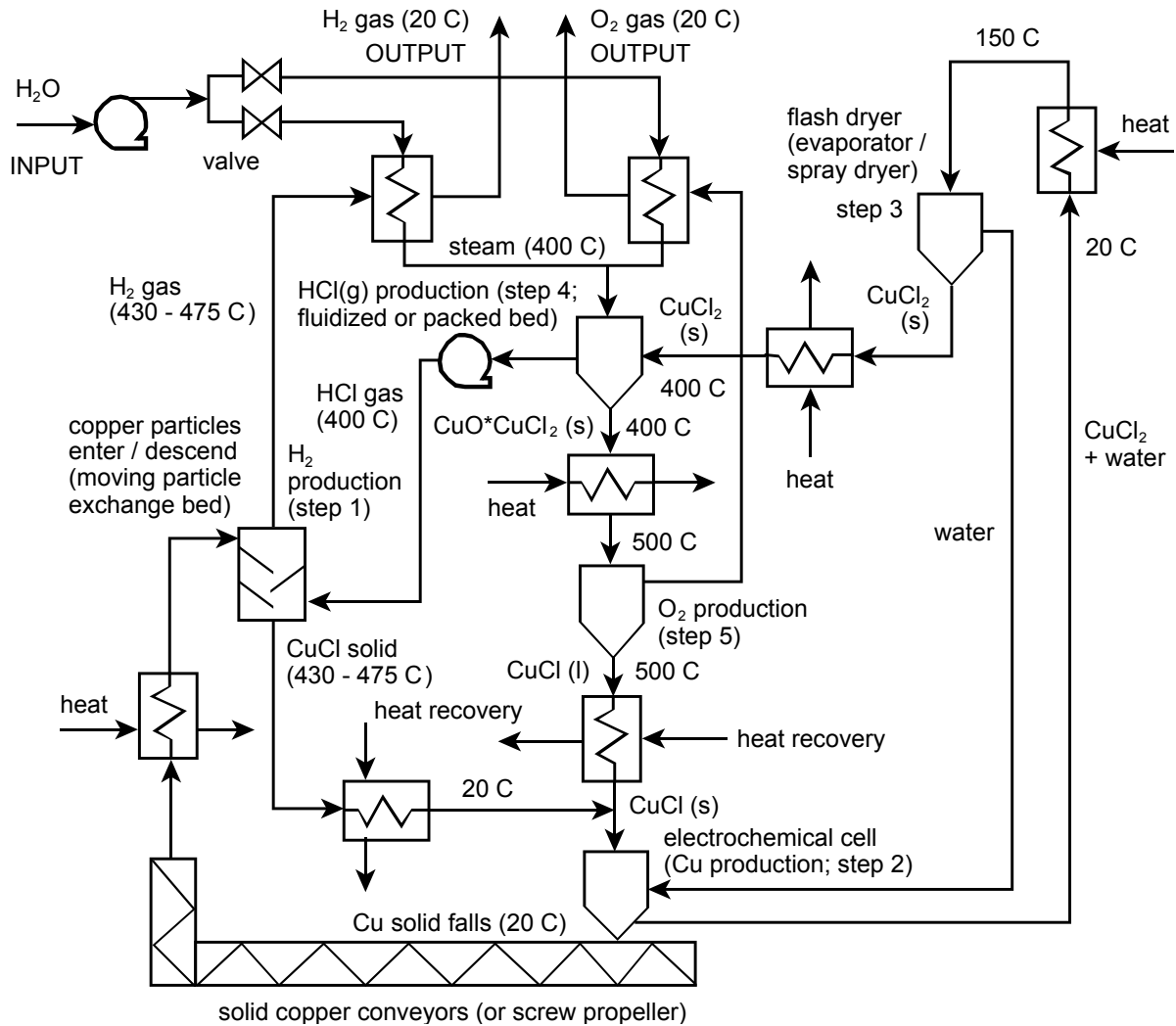


Figure 1: Conceptual layout of a Cu-Cl cycle

Table 1: Steps and chemical reactions in the Cu-Cl cycle

Step	Reaction	Temp. Range (°C)	Feed/Output*	
1	$2\text{Cu(s)} + 2\text{HCl(g)} \rightarrow 2\text{CuCl(l)} + \text{H}_2\text{(g)}$	430 - 475	Feed:	Electrolytic Cu + dry HCl + Q
			Output:	H ₂ + CuCl(l) salt
2	$2\text{CuCl(s)} \rightarrow 2\text{CuCl(aq)} \rightarrow \text{CuCl}_2\text{(aq)} + \text{Cu(s)}$	Ambient (electrolysis)	Feed:	Powder/granular CuCl and HCl + V
			Output:	Electrolytic Cu and slurry containing HCl and CuCl ₂
3	$\text{CuCl}_2\text{(aq)} \rightarrow \text{CuCl}_2\text{(s)}$	>100	Feed:	Slurry containing HCl and CuCl ₂ + Q
			Output:	Granular CuCl ₂ + H ₂ O/HCl vapours
4	$2\text{CuCl}_2\text{(s)} + \text{H}_2\text{O(g)} \rightarrow \text{CuO*CuCl}_2\text{(s)} + 2\text{HCl(g)}$	400	Feed:	Powder/granular CuCl ₂ + H ₂ O(g) + Q
			Output:	Powder/granular CuO*CuCl ₂ + 2HCl (g)
5	$\text{CuO*CuCl}_2\text{(s)} \rightarrow 2\text{CuCl(l)} + 1/2\text{O}_2\text{(g)}$	500	Feed:	Powder/granular CuO* CuCl ₂ (s) + Q
			Output:	Molten CuCl salt + oxygen

* Q = thermal energy, V = electrical energy

In the following analysis of heat requirements within the Cu-Cl cycle, stoichiometric relations will be assumed. Also, it is assumed that only water is consumed and then converted to hydrogen and oxygen, while other materials such as Cu, CuCl, CuCl₂ and HCl for cooling and heating are all recycled. The stoichiometric relation is 1 mol H₂O => 1mol H₂ + 1/2molO₂ or equivalently, 18g H₂O => 2g H₂ + 16g O₂. In the thermochemical cycle, there are some reactions and cooling processes that release heat. The total heat released from these reactions and cooling processes is calculated as $\Delta H_T^- = -290.11$ kJ, where the minus symbol “-” means the heat is “released” rather than “required”. The reactions and processes that *release* heat are listed as follows:

- H₂(g, 450 °C) → H₂(g, 25 °C) -4.22%
- 1/2O₂(g, 500 °C) → 1/2O₂(g, 25 °C) -2.54%
- 2Cu(s) + 2HCl(g) = 2CuCl(l) + H₂(g) 450 °C -16.14%
- 2CuCl(l, 450 °C) → 2CuCl(s, 25 °C) -27.87%
- 0.624H₂O (vapor, 100 °C) → 0.624 H₂O (liquid, 25 °C) -10.00%
- 2CuCl(l, 500 °C) → 2CuCl(s, 25 °C) -29.24%
- 0.624H₂O (vapor, 100 °C) → 0.624 H₂O (liquid, 25 °C) -10.00%

where the percentage means the ratio of the process heat (or reaction heat) to the total released heat.

If the released heat within the thermochemical cycle is not recycled internally, so no heat released by the reactions and cooling processes is used, then the total heat required by the reactions and cooling processes within the thermochemical cycle is $\Delta H_T^+ = 451.65$ kJ. The reactions and processes that *require* heat are listed as follows:

- 2Cu(s, 25 °C) → 2Cu(s, 450 °C) 5.18% (2)
- 0.624H₂O in Cu slurry (l, 25 °C) → vapor (g, 100 °C) 6.42% (3)
- CuO*CuCl₂(s) = 2CuCl(l) + 1/2O₂(g) at 500 °C 28.6% (4)
- 2CuCl₂(s) + H₂O(g) = CuO*CuCl₂(s) + 2HCl(g) 400 °C 25.82% (5)
- H₂O(l, 25 °C) → H₂O(g, 400 °C) 12.79% (6)
- 2CuCl₂(s, 25 °C) → 2CuCl₂(s, 400 °C) 12.00% (7)
- 0.624H₂O in CuCl₂ solution(l, 25 °C) → vapor (g, 100 °C) 6.42% (8)
- 4CuCl + 4Cl⁻ = 4CuCl₂⁻ in solution, 25 °C 0.06% (9)
- 4CuCl₂⁻ = 2CuCl₂ + 2Cu(s) + 4Cl⁻ in solution, 25 °C 2.71% (10)

where the percentage means the ratio of the process heat (or reaction heat) to the total required heat (not including the recycled heat released from the reactions and processes within the cycle).

The ratio of the released heat (ΔH_T^-) to the required heat (ΔH_T^+) is around 2/3, indicating the importance to recycle the released heat. However, this does not imply that recycling of released heat within the cycle is cost-effective, as some heat released within the cycle is low-grade heat (such as heat carried by low temperature water or the heat carried by solid powders). In engineering practice, this heat may be difficult to be recycled within the thermochemical cycle, or the cost to recycle this heat may be excessively high. As the quality of released heat from different reactions and processes are different, effectively transferring heat to different processes is an ongoing topic of thermal optimization.

In order to reduce the amount of required heat, a sensitivity analysis was performed for each chemical reaction and its heat requirements. In equation (2), the required heat is difficult to lower as the solid Cu is produced from electrolysis cell, which operates at ambient temperature. In the reaction using Cu as a reactant, the reaction temperature is fixed, which implies that Cu must be heated to this reaction temperature.

In equations (3) or (8), the heat required to dry H_2O is 87.9% due to evaporative heat transfer, which is difficult to reduce. In order to lower this heat flow, the main way is to lower the quantity of H_2O that must be dried. If 50% less liquid water forms in the slurry, the role of drying H_2O will decrease from 6.42% to 3.28%. However, to reduce the liquid water usually means increasing the cost of mechanically separating Cu or CuCl_2 from liquid water. Conversely, upon doubling the amount of liquid water, the role of drying H_2O will change from 6.42% to 11.38%.

The heat transfer in equations (4) and (5) is due to reaction heat, so it is difficult to lower. In the heat transfer in equation (6), 71% of the heat is represented evaporative heating, so this heat is difficult to lower. The water vapour could be provided directly from the exhaust vapour downstream of a low-pressure turbine that transfers mechanical energy to electricity in the nuclear station, which would lower the heat amount required within the Cu-Cl cycle. In equation (7), the heat is difficult to lower since the solid CuCl_2 is derived from the electrolysis cell, which is at the ambient temperature. In the reaction using CuCl_2 as a reactant, the reaction temperature is fixed, which means CuCl_2 must be heated to this reaction temperature. Also, the heat transfer in equations (9) and (10) is reaction heat, so it is difficult to lower. However, if the heat transfer rate is slower, then low-grade heat could be used in this process to heat the solution.

In general, to lower the amount of the required heat is difficult. One major reason for this difficulty is the heat required by the cycle mainly consists of reaction heat and latent heat (evaporation heat). These two forms of heat are determined by the thermodynamic properties of the copper-chlorine compounds, so it is difficult to lower these heating values, except if we change reactants and heating media. A possible way to lower the amount of required heat is to reduce the liquid water amount in the CuCl_2 and Cu slurry.

3. Heat Transfer in the Oxygen Reactor

The oxygen production step (step 5) receives solid feed of $\text{CuO} \cdot \text{CuCl}_2$ and produces O_2 gas and liquid cupric chloride. The melting point of CuCl is 430°C and the decomposition temperature of $\text{CuO} \cdot \text{CuCl}_2$ is higher than 450°C . Therefore, CuCl will leave the reactor in molten form. The heat carried by molten CuCl is high-grade heat. This heat can be recovered to heat reactants to the reaction temperature in various processes, such as: the reaction of Cu with HCl gas to produce hydrogen gas at a temperature of 450°C , the thermal energy carried by molten salt to heat the HCl to its reaction temperature, the reaction of CuCl_2 with water vapor to produce $\text{CuO} \cdot \text{CuCl}_2$ and HCl gas, or the heat carried by molten salt to heat the H_2O to a reaction temperature of 400°C . To recover the heat carried by molten salt, two types of contact methods between gas and molten salt are available: direct contact or indirect contact. To recover heat from molten CuCl by direct contact, the molten salt is sprayed downward and gas is flowing upward. This process of direct contact cooling involves a counter-current flow (illustrated in Fig. 2).

The heat carried by the molten salt can be used to heat the H_2O to a reaction temperature of 400°C in the reaction of CuCl_2 with water vapor to produce $\text{CuO} \cdot \text{CuCl}_2$ (step 4), rather heating HCl gas to a reaction temperature of 450°C in the reaction involving copper and HCl gas to produce hydrogen (reaction 1). The reasons are listed below as follows.

- The temperature of HCl produced in reaction 4 lies in the range of 400°C , which is closer to the temperature of molten CuCl , thereby indicating that the driving force of heat transfer will be smaller and it is difficult to make molten salt condense to solid particles. However, to heat H_2O has more choices of temperature, such as water vapor at 100°C or 200°C , including liquid water.
- Reaction 1 is exothermic, which means the HCl could be self-heated by the reaction heat.
- To heat HCl using CuCl means the equipment material must be HCl corrosion resistant. However, this problem does not arise with heating of H_2O .

- As CuCl goes to the solution of H₂O+HCl to undergo an eletrolysis process, this electrolysis process requires heat input. Therefore, to cool the CuCl particles using liquid water can directly prepare the solution and simultaneously provide heat to the solution.

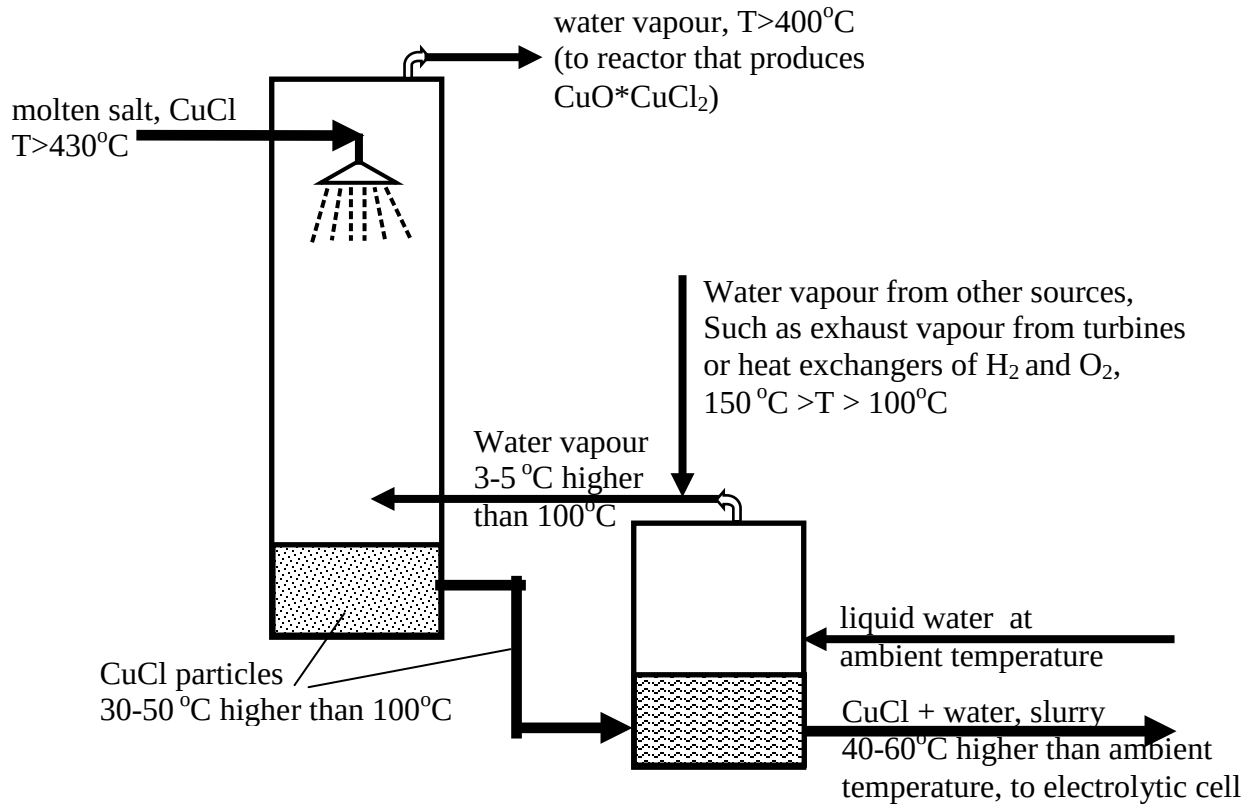


Figure 2: Cooling of molten salt by direct contact

The heat carried by the molten salt is enough to heat liquid water from 25°C to water vapor at 400°C (unsaturated). The heat in 2 mol CuCl(l) is assumed to be recovered from 450°C before entering the electrolytic cell with a CuCl + HCl + H₂O solution. As CuCl is cooled from liquid to solid, the pressure effect on the heat capacity and condensing heat can be neglected. During the cooling process from molten salt to solid, CuCl experiences β -hexagonal crystal ($412\text{--}423^{\circ}\text{C}$, denoted as β) and cubic ($<412^{\circ}\text{C}$, denoted as C) crystal forms. The heat released by 2 moles of CuCl(l) from liquid of 450°C to cubic solid of 25°C can be calculated as follows.

$$2(\Delta H_{\text{CuCl}})_{450-25} = 2(\Delta H_{\text{CuCl}})_{450,l} + 2(\Delta H_{\text{CuCl}})_{423,l} + 2(\Delta H_{\text{CuCl}})_{423,s,\beta} + 2(\Delta H_{\text{CuCl}})_{412,s,\beta} + 2(\Delta H_{\text{CuCl}})_{412,s,C} \quad (11)$$

where

$$(\Delta H_{\text{CuCl}})_{450,l}^{423,l} = \int_{450}^{423} C_p^{\text{CuCl},\text{liquid}} dT = -3.0 \quad \text{kJ} \quad (12)$$

$$[(\Delta H_{\text{CuCl}})_{423,l}^{423,s,\beta} + (\Delta H_{\text{CuCl}})_{423,s,\beta}^{412,s,\beta} + (\Delta H_{\text{CuCl}})_{412,s,\beta}^{412,s,C}] = -14.5 \text{ kJ} \quad (13)$$

$$(\Delta H_{\text{CuCl}})_{412, \text{solid}, C}^{25, \text{solid}, C} = \int_{412}^{25} C_P^{\text{CuCl}, \text{solid}, C} dT = -22.92 \quad \text{kJ} \quad (14)$$

Then we have $2(\Delta H_{\text{CuCl}})_{450, \text{liquid}}^{25, \text{solid}, C} = 2 \times 40.42 = 80.84 \quad \text{kJ}$. The ratio of the heat required by water to the heat released by molten CuCl is about 0.75. Also, the reaction to produce hydrogen also produces 2 moles of CuCl(l) in reaction 1, so the total quantity of molten CuCl will be 4 moles and the corresponding heat released by these 4 moles of CuCl will be 160kJ. Fortunately, the technology related to spraying and powder cooling towers is well established, for the operation of cooling molten salt using the direct contact between H₂O and molten CuCl. When heating HCl gas or H₂O vapor using the molten salt, some CuCl particles are likely to be entrained by HCl gas or H₂O vapour in the reactor, which will result in the slight decrease of contact area between reactant particles and reactant gas. This would result in a slight decrease of the reaction rate, as well as the product yield. However, in reaction 1, CuCl is a desirable product and in reaction 4, the entrained CuCl particles will mix with CuO*CuCl₂ particles that are consumed in reaction 5, in which CuCl is also a desirable product.

The second method of recovering heat from molten CuCl is by indirect contact (only suitable for T>430°C). The heat exchanger between gas and CuCl molten salt can be a traditional type of shell and tube heat exchanger. However, this indirect contact method can only be operated at a temperature higher than 430°C (melting point of CuCl), otherwise condensation of CuCl to solid in the pipe will plug equipment. If the temperature of CuCl has become equal or lower than 430°C, direct contact means must be adopted. Therefore, a two-step cooling process of molten CuCl could be adopted in order to recover the heat of molten salt. Step 1 would cool the molten CuCl to a temperature slightly higher than 430C in a heat exchanger, usually a traditional tube and shell heat exchanger. Step 2 must then use direct contact cooling as described earlier (spray cooling), otherwise pipes would be blocked.

In the removal of molten CuCl, reactant particles may be entrained by the effluent molten CuCl, because the effluent flow may induce a flow in molten CuCl in the reactor. In addition, fine oxygen bubbles will appear on the surface of the reactant particles when decomposing and if the bubbles detach slowly from the particles, it is likely that the reactant particles will not drop down or drop down very slowly. This would enhance the effluent entrainment of reactants, which should be avoided. Some bubbles may be entrained by the molten CuCl, so the bubbles may plug the pipe where the molten salt is flowing out of the reactor. The entrainment of reactant particles could also bring some safety issues involving the removal equipment. The particles may continue their decomposition in the pipe after leaving the reactor, which will generate gas bubbles in the pipe and simultaneously decrease the temperature of molten salt. This may result in solidification of molten salt since the decomposition requires heat.

There are various ways to avoid the entrainment. One way is to separate the reactor into two parts, using a plate or separator. Part 1 is mainly used for feeding reactants and the reaction, where it is better that all reactants are consumed. Part 2 is mainly used for removal of gas and molten salt products. This setup is illustrated in Fig. 3. Another possible technique to avoid the entrainment is to use a wider buffer or stabilizing section that could be designed in the upper part of the reactor. For example, the diameter of the upper part is twice the lower part and the molten salt is removed from the side wall of the upper part, while the reactant is fed from the lower part - not the bottom. To set a buffer part will not only decrease the upflow rate of particles to avoid entrainment, but also decrease the risk of splashing of molten salt by breakup of bubbles. Existing iron processing technology can be used to avoid entrainment in the reactor design. Figure 4 shows an illustration of the design, which combines both the separator and stabilizing parts. Figures 5 and 6 depict a more detailed conceptual layout of the thermochemical cycle, including details with heat matching in the hydrogen and oxygen cells.

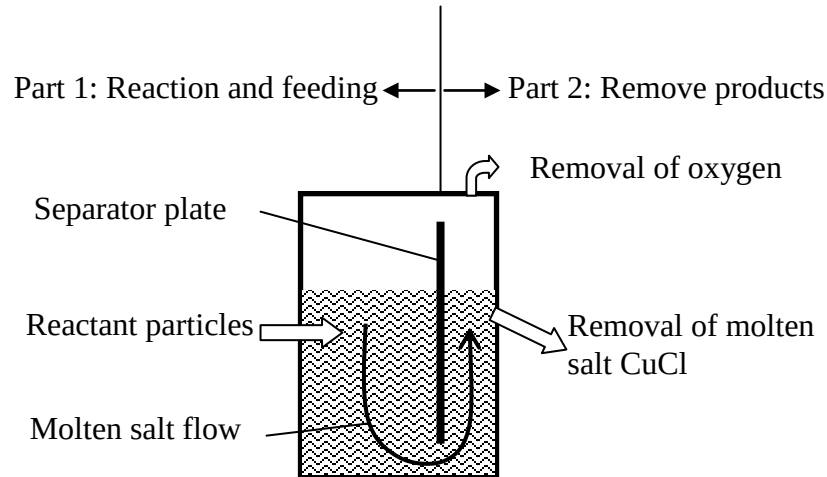


Figure 3: Schematic of molten salt vessel

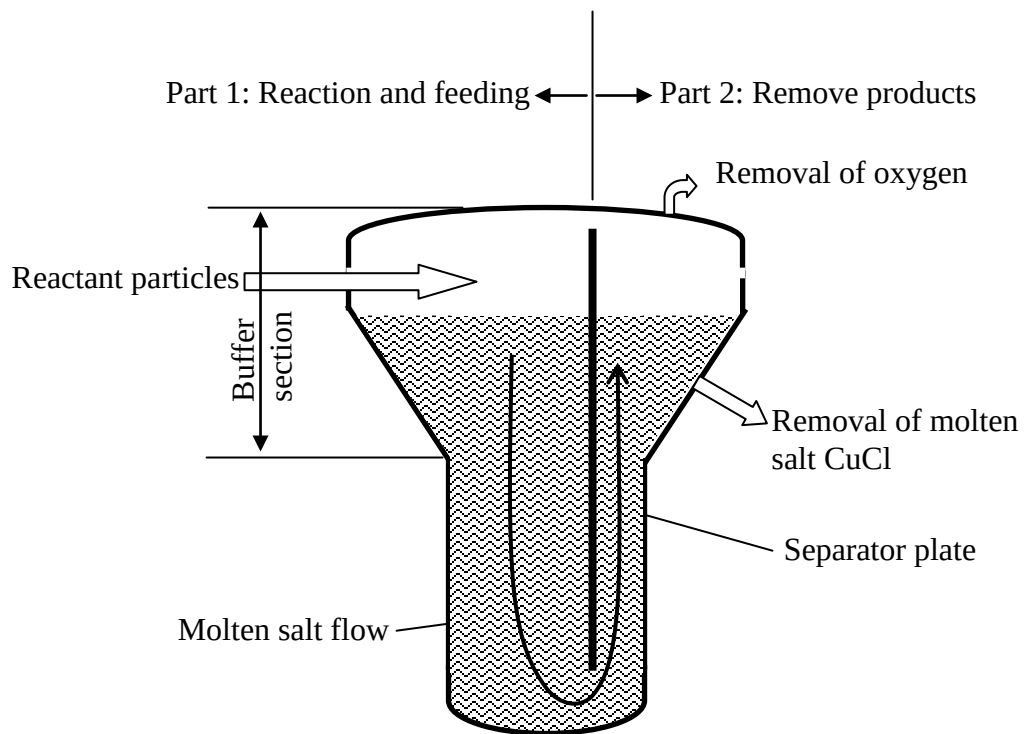


Figure 4: Alternative configuration with wider top to reduce entrainment

The reactant (copper oxychloride) will not melt because it must be first decomposed to oxygen and CuCl at 470 °C. The reactor to produce oxygen is operated at a temperature of 450 - 530°C. At this temperature, many metals would be oxidized by oxygen. The reactor and the oxygen removal equipment must be corrosion resistant to oxygen, molten CuCl and potentially chlorine gas. If CuCl₂ is entrained in the reactor with CuOCuCl₂, chlorine gas may appear and corrode the equipment material at the temperature of 530°C.

4. Conclusions

Between each step of the Cu-Cl plant, heat exchangers are needed for heat input or heat recovery and fluid transport to different portions of the thermochemical cycle. The oxygen production step has the highest temperature requirement (500 °C) in the cycle. Although the current CANDU reactors are unable to supply heat at such high temperatures, other transition alternatives (such as natural gas or hydrogen generated and recycled internally) can be utilized until the next generation of Canada's nuclear reactors, SCWR, becomes capable of producing the required high-grade heat.

This paper has specifically examined the heat requirements of the oxygen production and other steps within the Cu-Cl cycle. Heat supply is required at various temperatures throughout the cycle, so effective heat matching between endothermic and exothermic reactors is needed for efficient operation. Heat exchangers are typically located before and after each chemical reactor to supply fluid temperatures required for each reactor. This paper has examined the heat matching between steps of the Cu-Cl cycle, in efforts to recover as much heat as possible and minimize the net heat supply to the cycle, thereby improving its overall efficiency.

Acknowledgements

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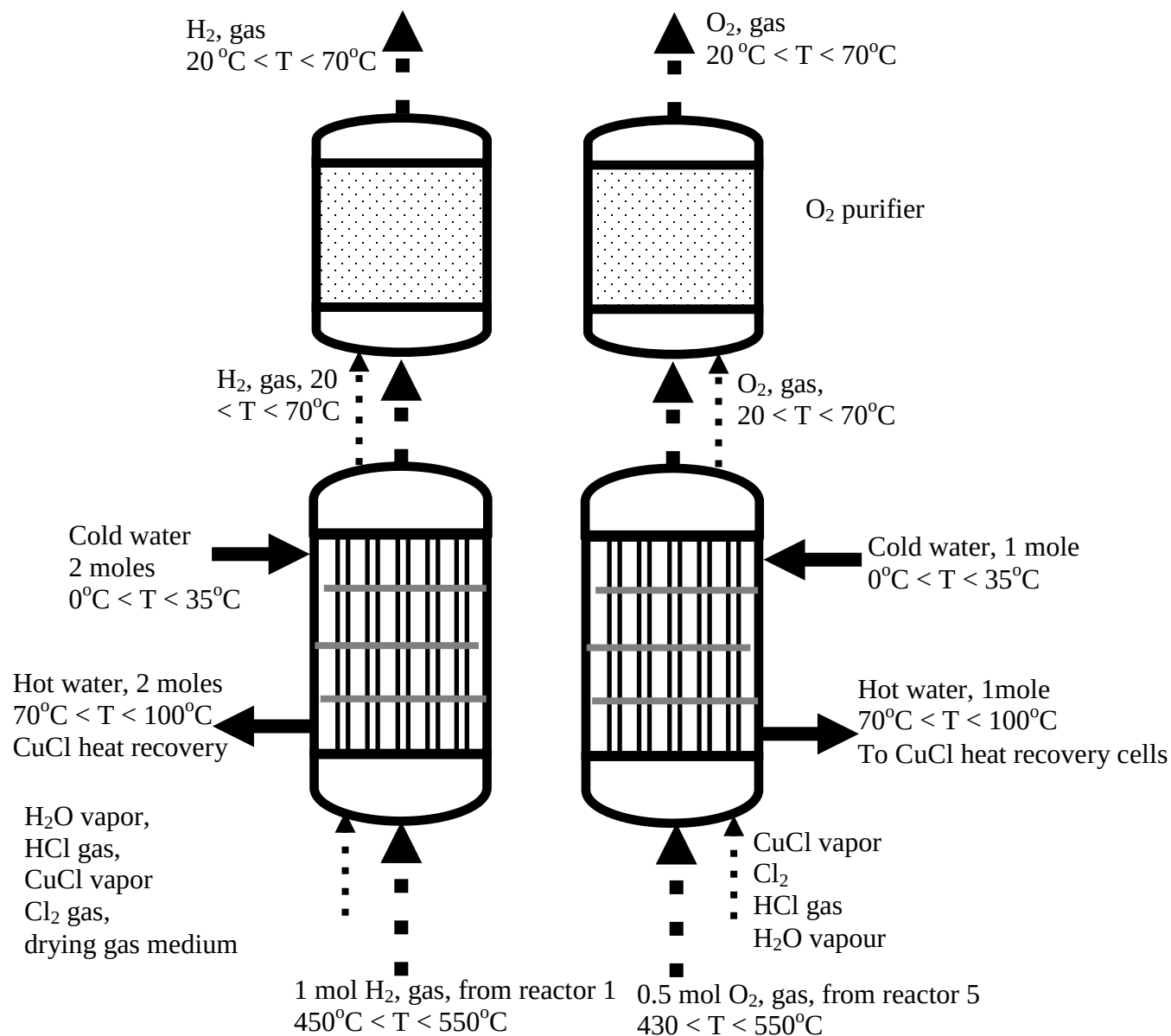


Figure 6: Heat recovery from hydrogen and oxygen cells